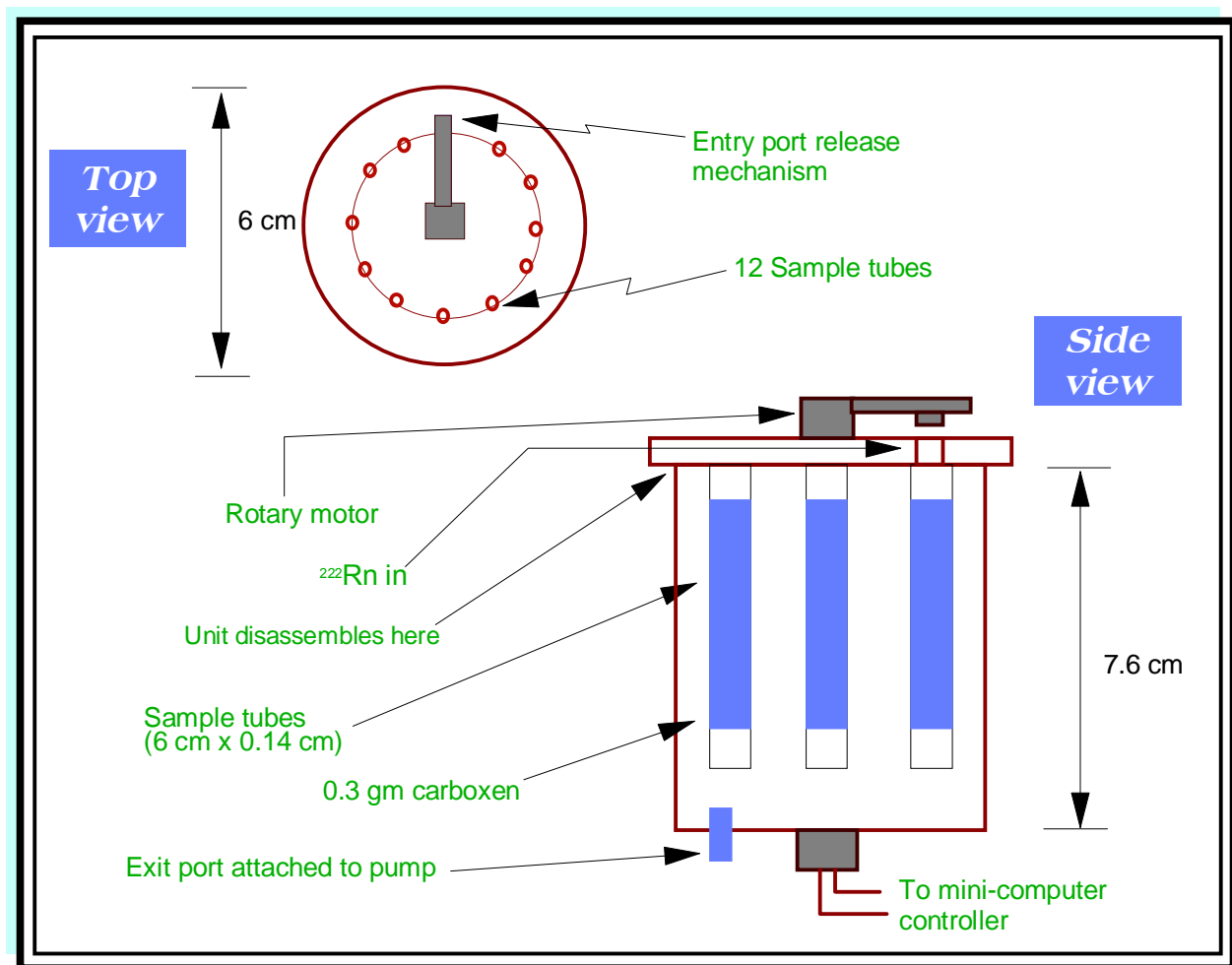


ENVIRONMENTAL MEASUREMENTS LABORATORY

EML-593



A Portable Battery-Powered Continuous Airborne ^{222}Rn Sampler



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April 1998

**A PORTABLE BATTERY-POWERED CONTINUOUS
AIRBORNE ²²²Rn SAMPLER**

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ABSTRACT

The Polyport, designed at the Environmental Measurements Laboratory (EML) for deployment in atmospheric balloons or remote areas, was laboratory and field tested to determine its effectiveness in collecting ^{222}Rn gas in dry and humid air. Twelve 6-cm long tubes containing 0.4 g of CarboxenTM-564 (20/45 mesh) are housed within the light-weight battery-powered Polyport unit. CarboxenTM-564 a hydrophobic beaded carbon molecular sieve (BCMS) material efficiently adsorbs airborne ^{222}Rn under dynamic sampling conditions of 1 - 2 h duration. The exposed sorbent is later weighed for water uptake, transferred and counted in a low background liquid scintillation (LS) counter that can detect alpha and beta emitting ^{222}Rn progeny with a maximum counting efficiency of 5 cpm per dpm. Each sorbent tube can be gamma counted if it contains sufficient adsorbed ^{214}Pb and ^{214}Bi activity. The variables tested were sampling flow rate, temperature, sampling time and relative humidity (RH). At standard temperature and pressure (STP) and an optimum sampling flow rate of 5 L h^{-1} , the sorbent's ^{222}Rn collection efficiency dropped from 58% to 15% as the RH increased from 20% to 80%. A single calibration curve was developed to estimate the ^{222}Rn collection efficiency, based on data obtained from the amounts of both ^{222}Rn and water vapor adsorbed by the BCMS material over a 20°C to 35°C temperature range. This calibration curve was used in two field tests and the hourly ^{222}Rn levels were compared with the levels obtained using the EML Radometer, an alternative ^{222}Rn measuring device. The minimum detectable ^{222}Rn concentration at STP for both a 1 h Polyport sample and a 1 h LS count was about 2 Bq m^{-3} (0.12 pCi L^{-1}) at 20% RH, based on an optimum sampling flow rate of 5 L h^{-1} , 58% collection efficiency, and 6 mBq lower limit of detection of the LS measurement.

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I NTRODUCTION

^{222}Rn , the decay product of ^{226}Ra , is a naturally occurring radioactive gas, whose short-lived progeny may pose a danger to human health (Nero 1983). Radon primarily enters the atmosphere after diffusing through the soil. Secondary sources include the ocean, ground water, natural gas, geothermal fluids and coal combustion (UNSCEAR 1988). In the atmosphere, ^{222}Rn decreases exponentially with increasing altitude over land with an outdoor air concentration of about 8 Bq m^{-3} at sea level and 2 Bq m^{-3} at 0.6 km above sea level (NCRP 1988).

Indoor and outdoor concentrations vary diurnally and seasonally. Indoor concentrations range from $11 - 300 \text{ Bq m}^{-3}$. The U.S. Environmental Protection Agency (USEPA) recommends that remedial action be taken in a home if ^{222}Rn concentrations exceed 150 Bq m^{-3} (4 pCi L^{-1}) (USEPA 1986).

Since both indoor and outdoor ^{222}Rn concentrations continually change with time, a collection device that provides continuous ^{222}Rn measurements and that is lightweight and inexpensive to operate may be advantageous. In this study, a portable 800 g battery-powered, constant-flow pump system packed with gram amounts of a hydrophobic beaded molecular sieve sorbent (BCMS; Betz et al. 1989) which has a high affinity for ^{222}Rn but not for water vapor was tested under dry and humid conditions. The small mass of sorbent may be counted for adsorbed ^{222}Rn progeny activity using either a NaI(Tl) gamma or conventional liquid scintillation (LS) counters.

A calibration curve was developed to account for the adverse effect of relative humidity (RH) on the sorbent's ^{222}Rn collection efficiency. The calibration curve was used in two field tests to determine the average hourly ^{222}Rn concentrations corrected for the amount of water vapor co-adsorbed by the sorbent material.

APPROACH

Without regard for temperature effects, the amount of ^{222}Rn passing through the sorbent tube, Q_{in} (Bq), depends on the sampling flow rate, SFR (L h^{-1}), sampling time, t (h), and the air ^{222}Rn concentration, C_o (Bq m^{-3}).

$$Q_{in} = \frac{SFR \times t \times C_o}{1000 \text{ L m}^{-3}} \quad (1)$$

The amount of ^{222}Rn collected by each tube, under specified conditions of RH, temperature and sampling time, is

$$Q_t = cph/Eff \quad (2)$$

where $Q_t = ^{222}\text{Rn}$ adsorbed by each tube (Bq) at t , $cph = \text{counts h}^{-1}$ determined by gamma or LS counting, $Eff = \text{instrument counting efficiency for } ^{222}\text{Rn progeny, } ^{214}\text{Pb, } ^{214}\text{Bi}$ ($\text{counts h}^{-1} \text{ Bq}^{-1}$). Q_t was determined by counting the sorbent material with either a gamma or LS counter, calibrated against a ^{226}Ra source of known activity. The percent ^{222}Rn % collection efficiency (CE) is the ratio of eqn 2 to eqn 1

$$^{222}\text{Rn \% CE} = [Q_t / Q_{in}] \times 100 \quad (3)$$

The mean air ^{222}Rn concentration, $\overline{C_o}$, is from eqns 1 - 3:

$$\overline{C_o} = \frac{1000 Q_t}{SFR \times t \times \%CE} \quad (4)$$

This simplified approach assumes a small temperature effect (see the Appendix) and requires a single calibration curve to describe the ^{222}Rn CE against the amount of co-adsorbed water vapor.

MATERIALS AND METHODS

A 0.8 kg (27 oz) constant airflow pump system (Polyport), designed at the Environmental Measurements Laboratory (EML), was used in this study. The Polyport (Polito and Albert 1994) is controlled by an onboard minicomputer that records surrounding temperature, RH and atmospheric pressure, and it also controls sampling times.

Twelve individual glass tubes are housed within the Polyport sampler. Each 6 cm long x 0.14 cm in diameter glass tube contains 0.4 g of CarboxenTM-564 (20/45 mesh),* a BCMS retained by wire mesh screens (see Fig. 1). CarboxenTM-564 is a spherical carbon molecular sieve material (particle diameter 354 to 841 μm) having a surface area of $400 \times 10^3 \text{ m}^2 \text{ kg}^{-1}$, and a density of $0.6 \times 10^3 \text{ kg m}^{-3}$. Its hydrophobic properties make it superior to activated charcoal, a common material used for adsorbing ²²²Rn. The BCMS material adsorbs four times less water than activated charcoal and retains twice as much ²²²Rn as 17 other charcoal types tested (Scarpitta 1992, 1995, 1996).

A rotary motor is attached to the entry port release mechanism of the Polyport that individually opens each valve above each sampling tube, allowing air to be pumped through by a 43 g pump.** The sampling flow rate, which could be adjusted from 4 to 8 L h⁻¹ for any preset time, was monitored at the exit port of the Polyport using a Brooks E/C meter. Less than 1% of the outside air leaks into the Polyport when it is sealed and fully packed with sorbent over a 48-h period. Following exposure, the Polyport is disassembled, each tube capped and then individually measured for both ²²²Rn and water vapor content.

The BCMS material was gamma counted for 15 to 30 min or quickly transferred to 20 mL low ⁴⁰K borosilicate glass vials containing 15 mL of chilled Perma-Fluor-V*** cocktail and securely capped. For gamma counting, the amount of ²²²Rn adsorbed by the sorbent (counts min⁻¹ kg⁻¹) was measured using an 8-cm diameter shielded Harshaw NaI (Tl) crystal interfaced to a Norland INO-Tec⁺ 5300 multichannel analyzer preset at 200 to 700 keV to measure the gamma peaks of the ²²²Rn progeny, ²¹⁴Pb and ²¹⁴Bi. For LS counting measurements, a Packard Tri-Carb 2250-CA LS analyzer was used with preset window settings of 50 to 1000 keV.

The background corrected counts obtained over the counting interval were converted to adsorbed ²²²Rn activity, Q_t (Bq), using the Countess (1976) equation. The Countess equation, which requires that the instrument detection efficiency be known, corrects for radioactive decay

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that occurs during sampling (typically 1 h), progeny ingrowth (3 h), and counting times (typically 1 h).

The lower level of detection (LLD) for each counting method was determined by taking multiple instrument background measurements (NCRP 1988). The LLDs at the 95% confidence limit for the gamma and the LS counters were 400 mBq (10.8 pCi) for a 0.5-h count, and 6 mBq (0.16 pCi) for a 1-h count, respectively.

The amount of water vapor adsorbed on the BCMS material was determined using a Sartorius balance. Weights of each preloaded tube were recorded after heating at 100°C and following ²²²Rn exposure, with the weight differences corresponding to water uptake, W (g H₂O kg⁻¹ sorbent) during exposure.

Separate experiments were conducted to determine both the optimum sampling time and sampling flow rate. Individual tubes containing BCMS material were connected to either a Brailsford TD-4×2 pump that could deliver 7 to 20 L h⁻¹ or a Gillian pump that could deliver 1 to 10 L h⁻¹. The sampling times ranged from 1 min to 2 h. All pumps were calibrated using a Gilian PN-D-800266 bubble generator. Calibration experiments were conducted in EML's 30 m³ walk-in Radon, Thoron, and Progeny Exposure Facility where conditions of temperature can be controlled from 5°C to 40°C and ²²²Rn levels from 100 to 5000 Bq m⁻³ (Fisenne and Cavallo 1997). The conditions can be programmed and controlled in 1-h increments for up to 2 weeks. During later field testing, a calibration curve, established from previous ²²²Rn chamber exposures, was used to determine the mean hourly ²²²Rn levels. An independent ²²²Rn measuring device, the Radometer (Negro 1990), was used in two field tests to verify the results obtained with the Polyport sampler. At 20% RH, the ²²²Rn detection efficiency of the Radometer is about 25% (0.04 counts min⁻¹ per Bq m⁻³) and drops to 20% at 70% RH. The Radometer estimates the ²²²Rn levels in air to within 10% of the true value at concentrations exceeding 100 Bq m⁻³.

RESULTS

SAMPLING TIME AND FLOW RATE

Fig. 2 shows the ²²²Rn collection efficiency using 6 cm BCMS sorbent tubes connected by Tygon tubing to either a Brailsford or Gillian pump at flow rates ranging from 1.5 to 10 L h⁻¹. Using eqn 1, the amount of ²²²Rn entering each tube was held constant at 3.7 Bq by varying the sampling time in a ²²²Rn environment of 1800 ± 36 Bq m⁻³. This was done to prevent saturation of the sorbent with ²²²Rn as the flow rate was increased. The data show that the ²²²Rn collection efficiency (~ 45%) was unaffected by increasing sampling flow rate when the amount of ²²²Rn entering each tube was held constant. The amount of water vapor adsorbed per tube was constant (10 to 12 g kg⁻¹ of sorbent) at 26% RH and T = 21°C.

In a separate experiment, the ^{222}Rn level in the chamber was maintained at 100 Bq m^{-3} with sampling times ranging from 1 min to 2 h at a constant sampling flow rate of 5 L h^{-1} . Fig. 3 shows that the optimum sampling time was about 1 h, based on the amount of adsorbed ^{222}Rn (mBq). An increase in the flow rate to 10 L h^{-1} did not affect the amount of ^{222}Rn adsorbed (data not shown). Based on these results, it was determined that a 1 h sampling time and a sampling flow rate of about 5 L h^{-1} be used throughout this study.

TEMPERATURE

Fig. 4 shows (a) the ^{222}Rn conditions in the chamber, and (b) the amount of ^{222}Rn and water vapor collected by each of six BCMS tubes that were exposed in the EML ^{222}Rn chamber at temperatures that ranged from 12.1°C to 32.0°C . The amount of ^{222}Rn (mBq) collected by each tube increased in response to increased ^{222}Rn levels in the chamber. However, the RH, which was not constant, had decreased from 40% - 20% during the exposure interval, resulting in continually decreasing water uptake by each successive tube within the Polyport. The ^{222}Rn % collection efficiency appeared to increase 13% over the experimental temperature range. For every degree Centigrade increase in temperature above 12°C , the ^{222}Rn collection efficiency increased by 0.6% due primarily to reduced water uptake with some contribution by increasing temperature. The temperature/humidity effect was later taken into account in the final calibration curve.

RELATIVE HUMIDITY

Figs. 5a and 5b show the amounts of both ^{222}Rn and water vapor collected by the 12 tubes contained within the Polyport while the RH changed from 25% to 80%. The temperature was maintained between 25°C and 28°C while the chamber ^{222}Rn levels remained relatively constant, ranging between 60 and 85 Bq m^{-3} . Radon adsorption by the BCMS material was highest when the RH was lowest and vice versa. A fourfold increase in the RH (20% to 80%) adversely affected the ^{222}Rn adsorption by dramatically reducing the collection efficiency from 56% to 15%, a 70% change.

CALIBRATION CURVE

The use of eqn 4 to estimate the average hourly ^{222}Rn concentration in air requires that a calibration curve be devised to correct the ^{222}Rn collection efficiency for water uptake and

temperature. Four independent experiments were done over a range of temperatures (20°C to 35°C) and varying RH to establish a single calibration curve for water uptake vs. ²²²Rn collection efficiency. The data in Fig. 6a show an inverse linear relationship between ²²²Rn and water uptake, as expected. There may be a slight temperature effect, where the % collection efficiency is diminished somewhat by the higher temperatures.

Linear regression of the data in Fig. 6a shows that the slope is -0.4 % collection efficiency per g H₂O kg⁻¹ of sorbent over a temperature range of 20°C to 35°C, with an intercept value of 58%. The equation describing the ²²²Rn collection efficiency of the BCMS material over a 20°C to 35°C temperature range at a sampling flow rate of 5 L h⁻¹, versus water uptake is

$$\% \text{ collection efficiency} = (-0.4 \times W) + 58\% \quad (5a)$$

where W is the amount of water adsorbed by the sorbent, g of H₂O kg⁻¹ of sorbent. The data in Fig. 6b (N = 40 and R² = 0.893) is a log-log plot showing the amount of water vapor, W, adsorbed by the sorbent as a function of the air water vapor content, C_w (g H₂O m⁻³ of air). Values for C_w at a given temperature and RH were obtained from the Steam Tables in Handbook of Physics and Chemistry (see the Appendix). In Fig. 6b, the amount of adsorbed water, W, as a function of C_w, is described by the following equation

$$W \text{ (g of H}_2\text{O kg}^{-1} \text{ of sorbent)} = 1.25 C_w^{1.4} \quad (5b)$$

Substituting eqn 5b into 5a yields

$$\% \text{ collection efficiency} = 58\% - [0.5 C_w^{1.4}] \quad (5c)$$

for a 1 h sample using 0.4 g of CarboxenTM-564 at a sampling flow rate of 5 to 6 L h⁻¹.

Eqns 5a through 5c were verified in a separate experiment by exposing the Polyport sampler in the EML chamber to a ²²²Rn concentration of 89 ± 12 Bq m⁻³ at 22°C and 22% RH. From eqn A1, C_w = 4.37 g of H₂O m⁻³ of air (see the Appendix). Using this value in eqn 5b, the expected amount of W is 9.85 g of H₂O kg⁻¹ of sorbent, slightly above the 1 standard deviation (SD) of the average measured value of 7.6 ± 22% g of water H₂O kg⁻¹ of sorbent. Based on the average amount of ²²²Rn adsorbed by each of the 12 tubes (Q_t = 0.283 ± 0.034% Bq) at a sampling flow rate of 6 L h⁻¹, the average hourly % collection efficiency was calculated as 55% using the actual value of W in eqn 5a, and 54% using C_w = 4.37 in eqn 5c. The average hourly

ambient ^{222}Rn concentration, C_o , using eqn 4, was 0.964 ± 0.093 of the true mean ^{222}Rn chamber value when the actual amount of W in eqn 5a was used to estimate the % collection efficiency.

FIELD TESTS

Two field tests were conducted at EML to determine how accurately the Polyport sampler estimated the levels of ^{222}Rn in dry and humid air. The first was conducted in a 5th floor office room at 64% RH, while the second test was conducted in the EML basement at 20% RH. In both tests, the Radometer was used to verify the hourly results obtained using the Polyport sampler. Eqn 5a was used to determine the ^{222}Rn % collection efficiency, based on the amount of water adsorbed by each tube. The radon concentrations were then determined using eqn 4.

In Table 1, the mean of the hourly measurements made using both the Polyport and Radometer at 64% RH were $5.8 \pm 2.6 \text{ Bq m}^{-3}$ and $8.1 \pm 3.0 \text{ Bq m}^{-3}$, respectively. The Polyport results agreed with a previous study by Fisenne (1994) where the average annual ^{222}Rn level in the test room was $6.6 \pm 10\% \text{ Bq m}^{-3}$.

In Table 2, the means of the hourly Polyport and Radometer measurements for the second test at 20% RH were $42.3 \pm 18.5 \text{ Bq m}^{-3}$ and $32.3 \pm 5.4 \text{ Bq m}^{-3}$, respectively.

CONCLUSION

A light weight, portable, battery-powered, constant flow pump system (Polyport) was tested to determine its effectiveness in collecting ^{222}Rn gas in humid air. Four variables were tested: (1) flow rate, (2) sampling time, (3) temperature, and (4) the RH. Based on independent experiments, it was determined that the optimum sampling time was 1 h when a sampling flow rate of 5 to 6 L h^{-1} was used. Any temperature effect between 12°C and 35°C was masked by water uptake. The determining factor in ^{222}Rn adsorption by the BCMS material was absolute humidity. Increasing the RH, from 20% to 80%, adversely affected the ^{222}Rn adsorption by reducing the ^{222}Rn collection efficiency by 70% at STP.

A single calibration curve was established to adjust the ^{222}Rn collection efficiency for water uptake over a temperature range of 12°C to 35°C . This approach estimated the air ^{222}Rn levels in two field tests to within 40% of the expected 1 standard deviation value, based on the Radometer, a secondary ^{222}Rn measuring device. In a high RH office with an average annual ^{222}Rn concentration of $6.6 \pm 10\% \text{ Bq m}^{-3}$ (Fisenne 1994), the mean hourly ^{222}Rn concentrations were $5.8 \pm 2.6 \text{ Bq m}^{-3}$ and $8.1 \pm 3.0 \text{ Bq m}^{-3}$ using the Polyport and Radometer, respectively. In a low RH basement, the mean ^{222}Rn level, using the Polyport was $32.3 \pm 5.4 \text{ Bq m}^{-3}$, whereas that of the Radometer was $29.9 \pm 9.9 \text{ Bq m}^{-3}$.

The Polyport, sampling at 5 to 6 L h⁻¹, can provide continuous, hourly measurements with a ²²²Rn collection efficiency of 58% in dry air (<20% RH) at STP. In humid air (80% RH) the collection efficiency dropped to about 15% at the same sampling flow rate. When LS counting is employed, the LLD for a 1 h count at the 95% confidence level was 6 mBq. Under the worse case conditions (i.e., very humid air), each tube in the Polyport can collect about 8 mBq of ²²²Rn h⁻¹ when the average ambient ²²²Rn concentration is 9 Bq m⁻³ at STP and collection efficiency = 15% (see eqn 4). Similarly, the Polyport should collect the same amount of ²²²Rn in dry air at STP, as compared with that in humid air, if the average ambient ²²²Rn concentration is 2 Bq m⁻³ and the collection efficiency = 58%. If flown on a balloon, this lightweight device could be useful in atmospheric climate studies where the ²²²Rn, humidity and temperature decrease with increasing altitude. Multiple units can be deployed in remote areas without a power supply or throughout a building, providing hourly ²²²Rn concentrations at environmental levels.

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TABLE 1

FIELD TEST IN EML ROOM 527 (T = 22° C: RH = 64%)

Tube ID	H ₂ O uptake (g kg ⁻¹)	Q(t) mBq collected ^a	Polyport (Bq m ⁻³) ^b	Radometer (Bq m ⁻³)
A	112.0	3.2	4.8	8.3
B	82.0	6.1	4.8	10.7
C	87.0	4.8	4.1	13.9
D	80.0	7.7	5.9	11.3
E	85.0	7.5	6.3	4.5
F	91.0	6.9	6.3	5.6
G	35.0	3.5	1.6	6.8
H	101.0	9.8	11.1	6.8
I	113.0	2.0	3.1	10.1
J	126.0	2.9	7.6	5.6
K	108.0	6.3	8.5	5.1
Average	85.7	5.5	5.8	8.1
1 sigma	35.5	2.4	2.6	3.0

^a ²²²Rn concentration calculated using eqns 4 and 5.^b 1 sigma LS counting error < 4%.

TABLE 2

FIELD TEST IN EML BASEMENT (T = 25°C: RH = 20%)

Tube ID	H ₂ O uptake (g kg ⁻¹)	Q(t) mBq collected ^a	Polyport (Bq m ⁻³) ^b	Radometer (Bq m ⁻³)
A	33.0	67.2	30.0	33.6
B	40.0	63.8	30.4	25.9
C	43.0	122.4	60.0	25.5
D	40.0	166.6	79.3	24.4
E	32.0	152.0	67.2	30.3
F	31.0	106.0	46.5	32.9
G	33.0	89.0	39.7	41.8
H	29.0	72.6	31.3	37.4
I	29.0	64.0	27.6	34.0
J	28.0	59.7	25.5	36.3
K	28.0	67.0	28.6	33.7
Average	33.2	93.7	42.3	32.3
1 sigma	5.3	38.1	~5	5.4

^a ²²²Rn concentration calculated using eqns 4 and 5.^b 1 sigma LS counting error < 4%.

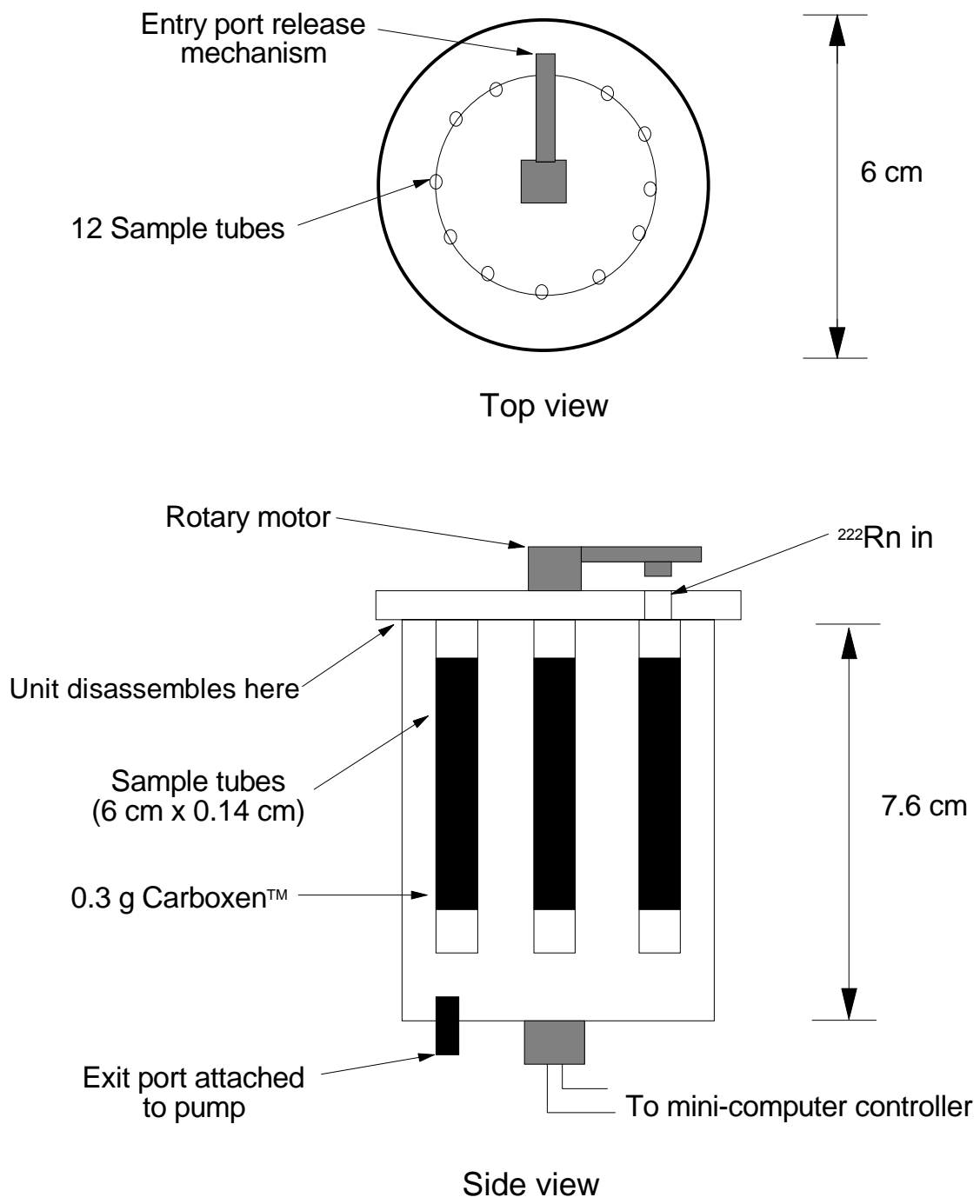


Figure 1. EML Polyport radon sampler (0.220 L inner volume).

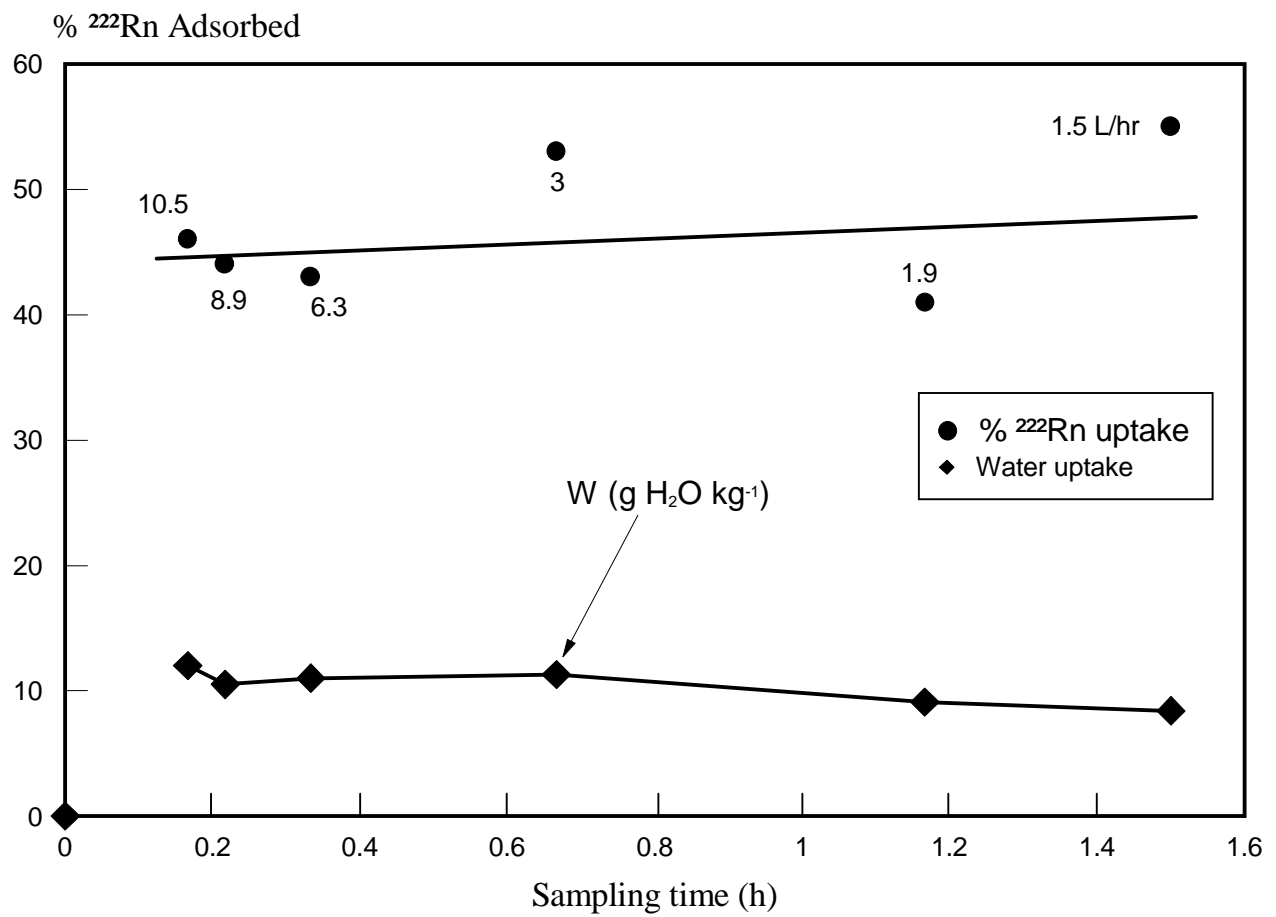


Figure 2. ²²²Rn collection efficiency by 6 cm of Carboxen™-564 (20/45 mesh) at various sample flow rates (1.5 - 10 L h⁻¹): Q_{in} = 3.7 Bq.

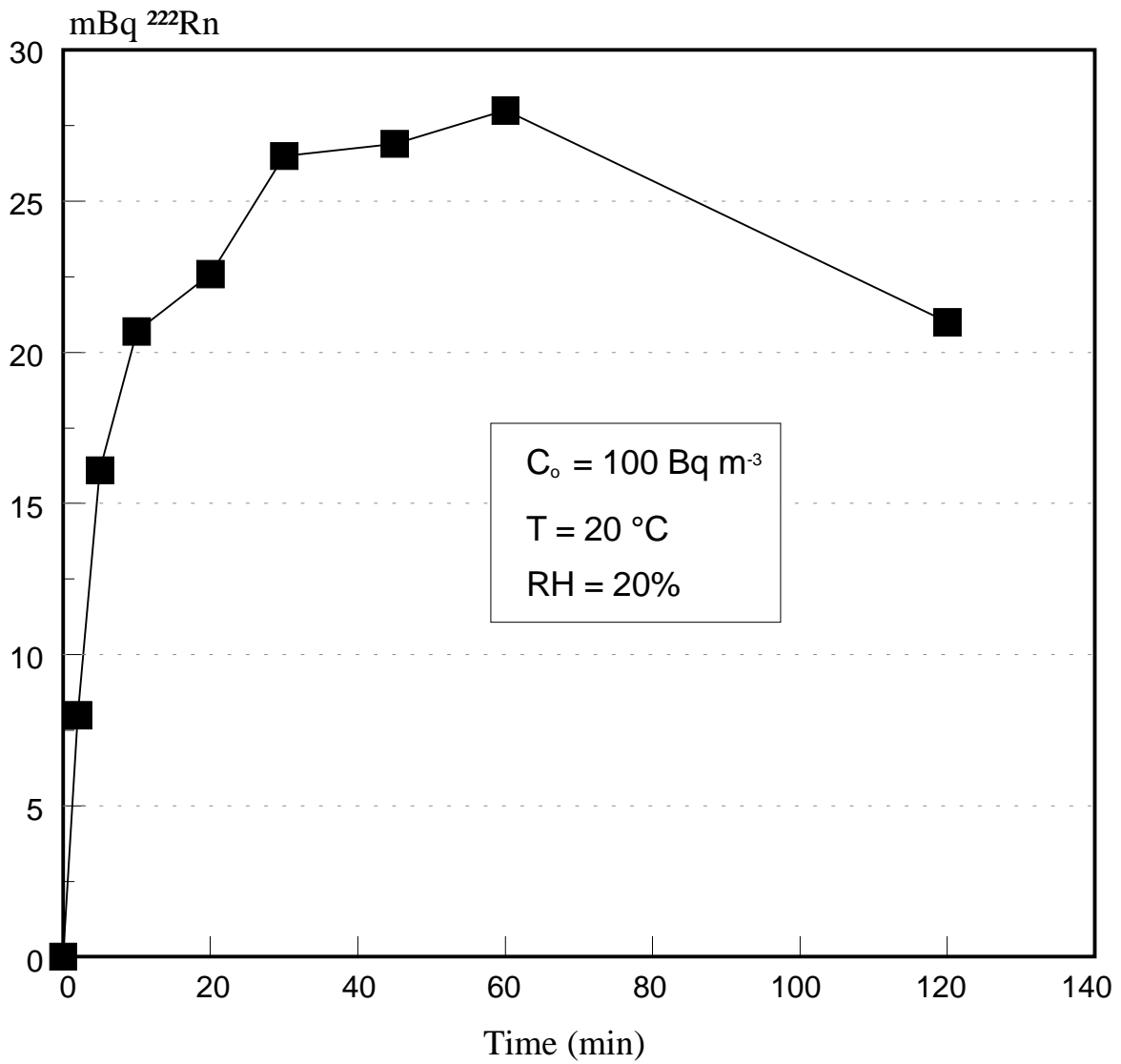


Figure 3. Optimum sampling time at sampling flow rate = 5 L h⁻¹.

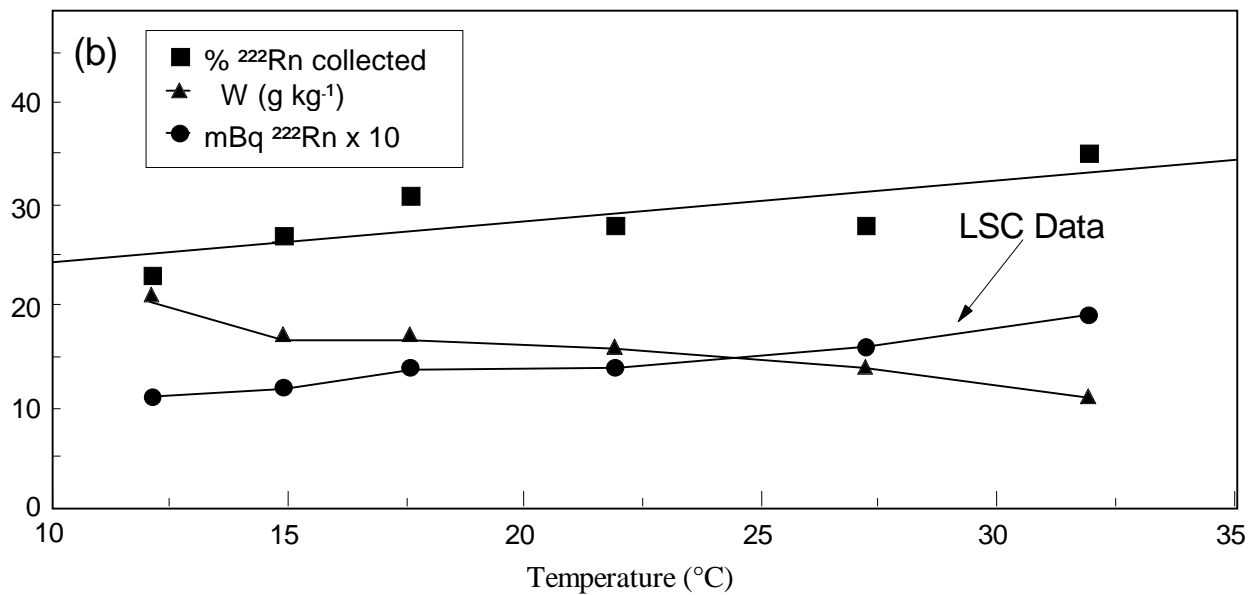
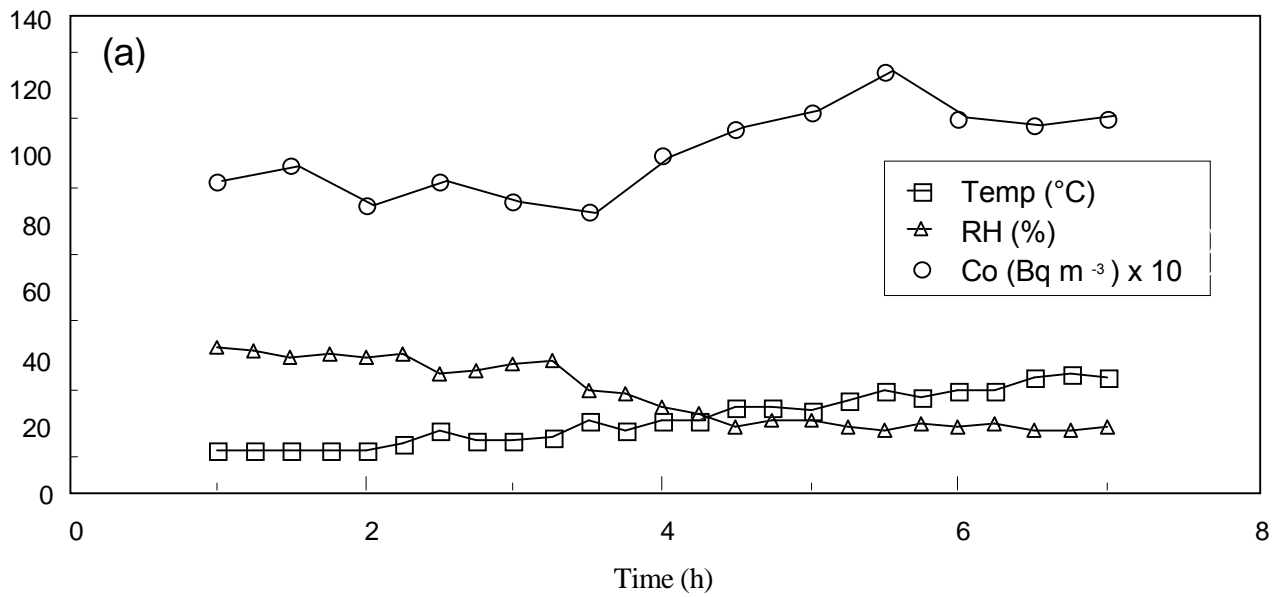


Figure 4. (a) EML ²²²Rn chamber exposure conditions;
 (b) Polyport response to changing temperature: sampling flow rate = 5 L h⁻¹ x 1 h per tube.

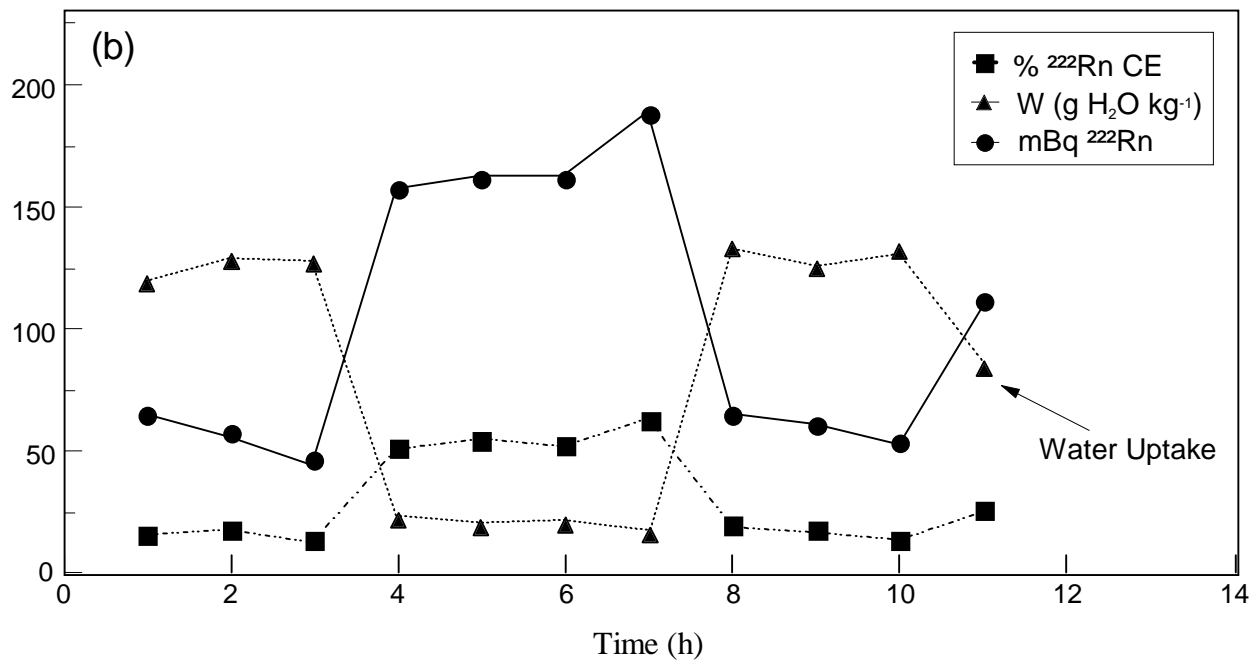
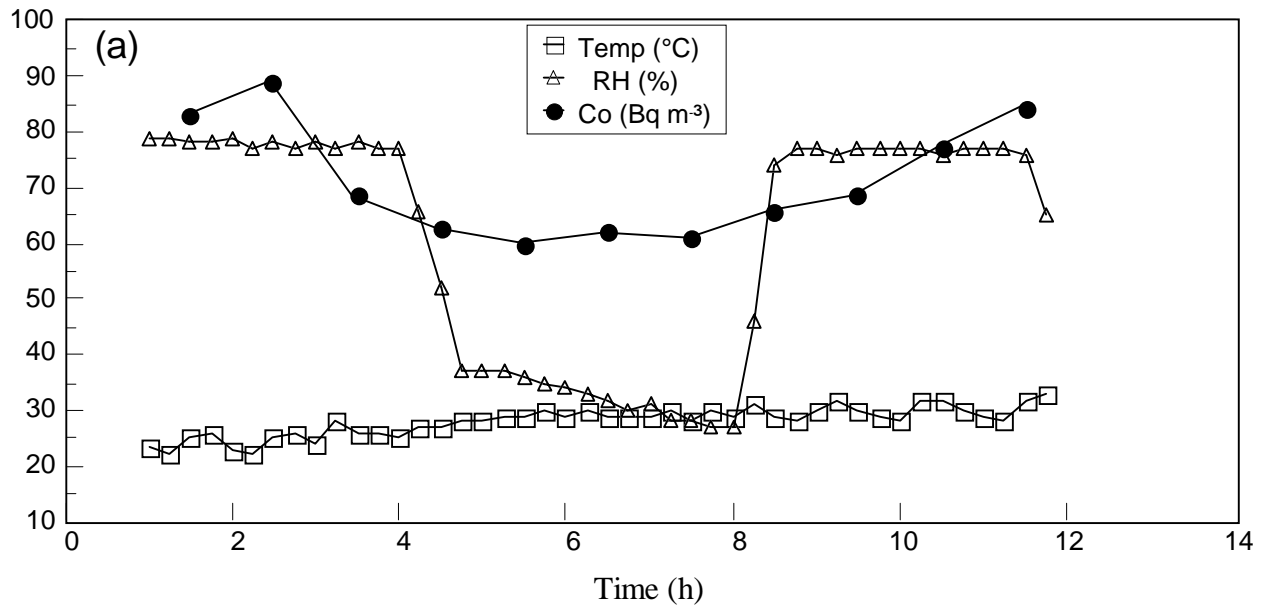
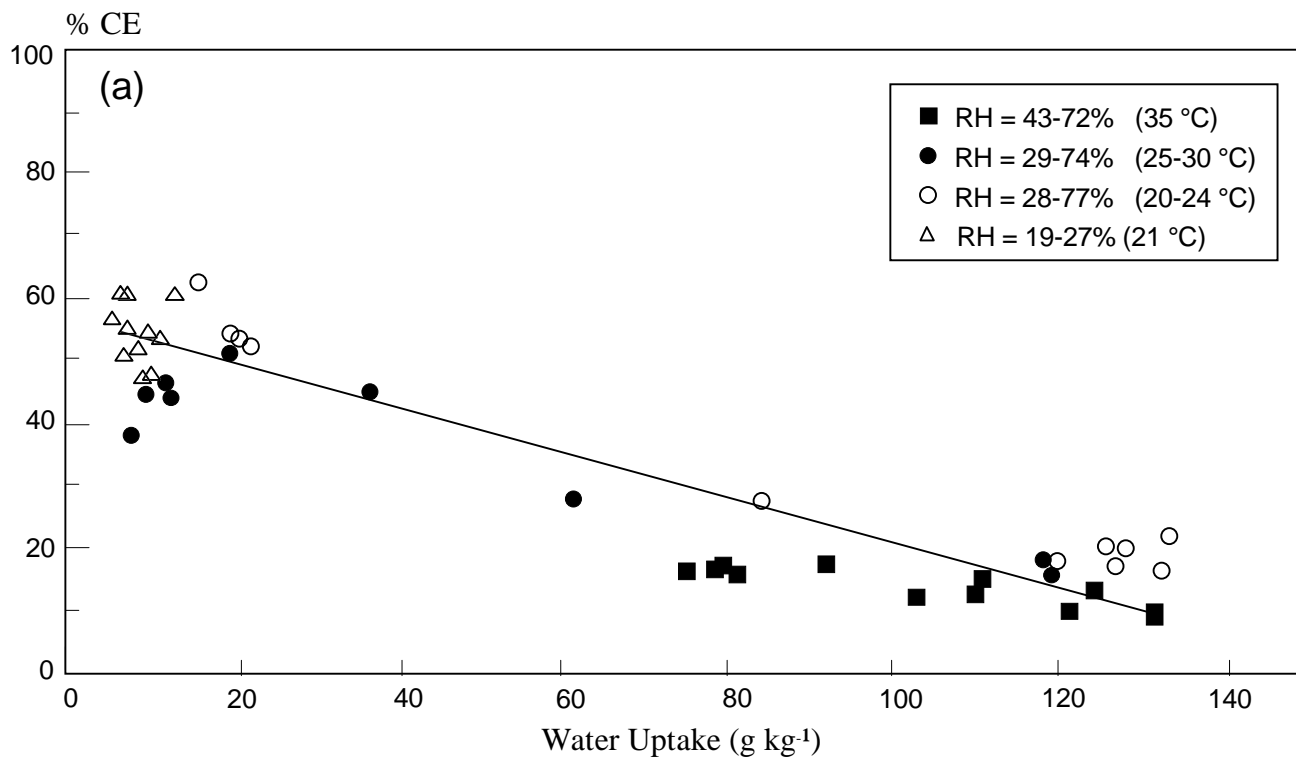


Figure 5. (a) EML ^{222}Rn chamber exposure conditions;
 (b) Polyport response to changing humidity: sampling flow rate = 5 L h^{-1} .



Air water vapor content, C_w (g m^{-3})

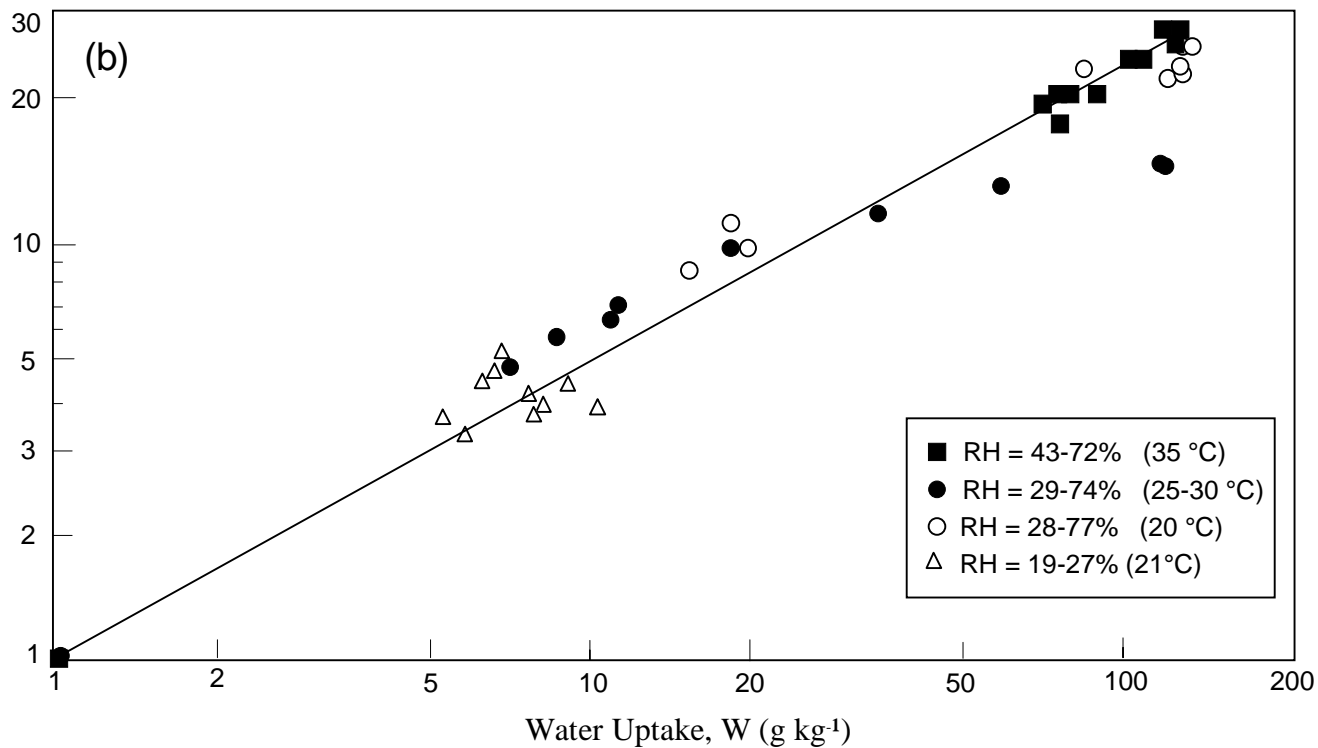


Figure 6. a) ^{222}Rn collection efficiency (CE) vs. H_2O uptake: sampling flow rate = 5 L h^{-1} ;
 b) Absolute humidity vs. sorbent water uptake: sampling flow rate = $5 \text{ L h}^{-1} \times 1 \text{ h}$.

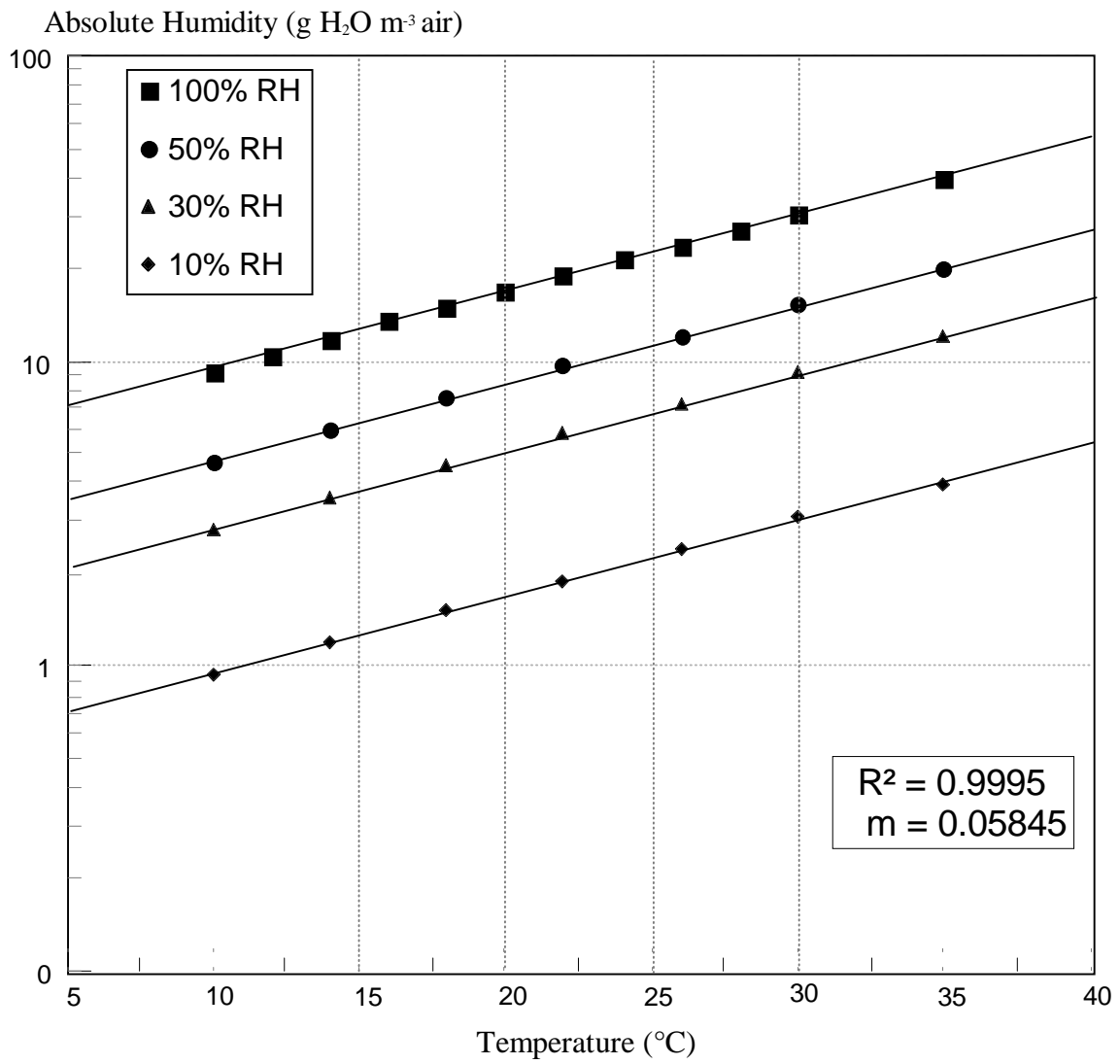


Figure 7. Absolute humidity vs. temperature.

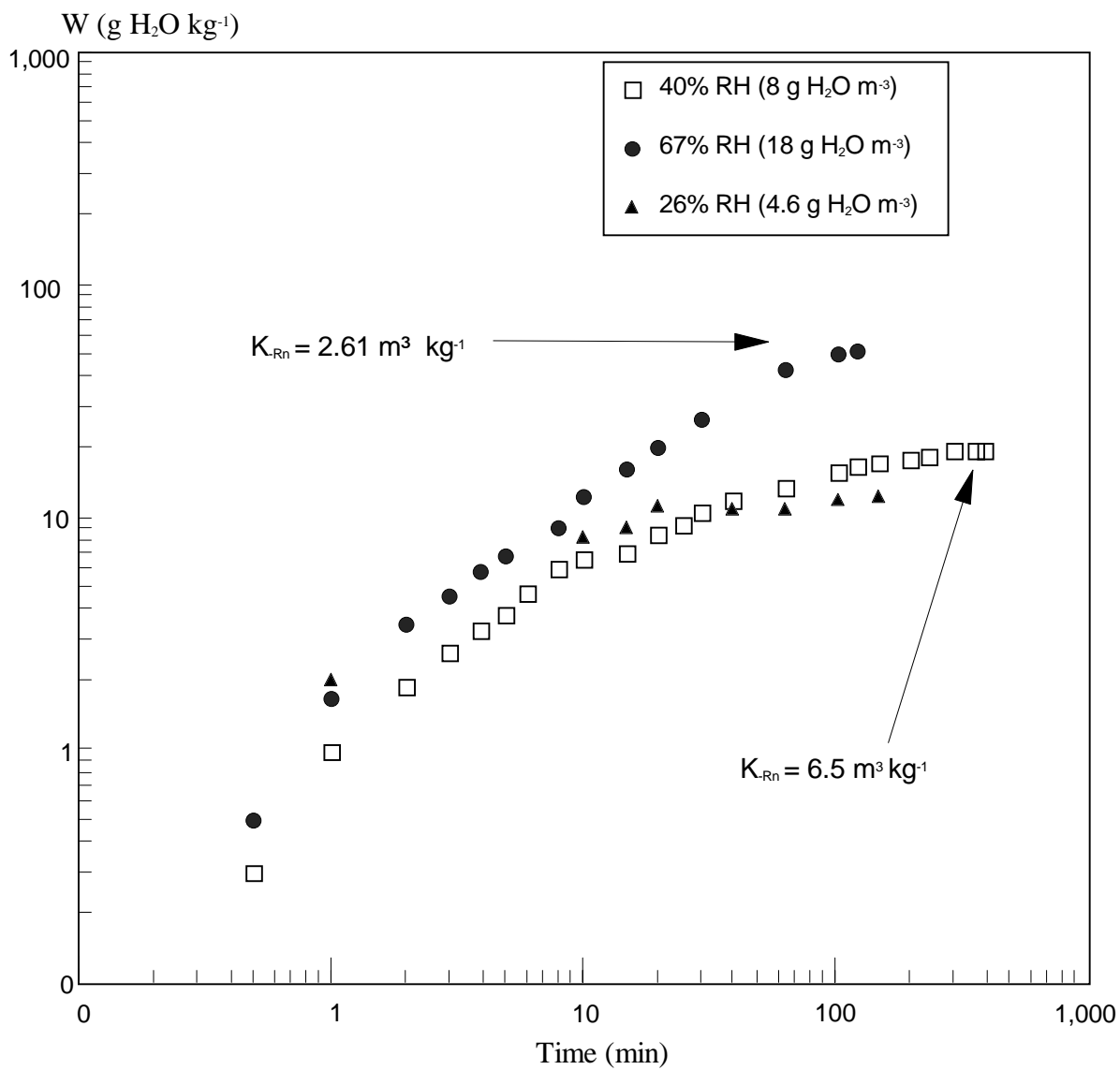


Figure 8. Water uptake by Carboxen™-564 in a thin layer at room temperature.

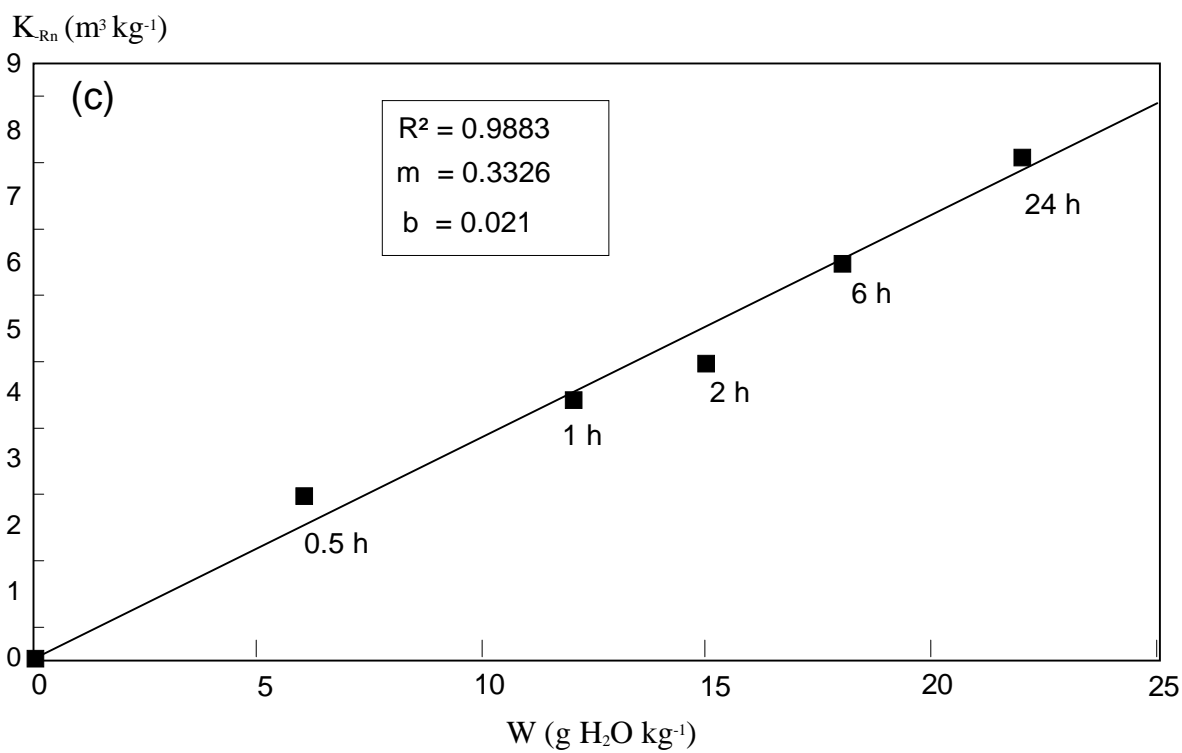
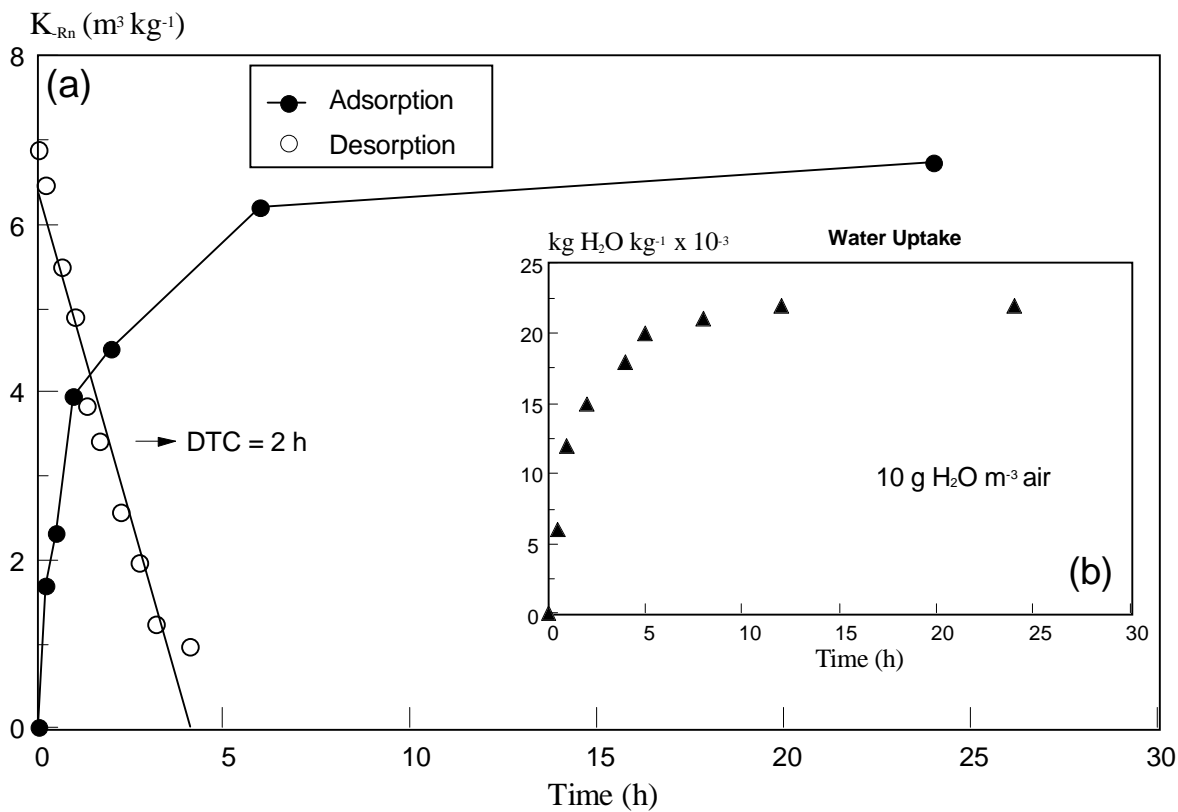


Figure 9a, b, c. ²²²Rn adsorption and desorption by Carboxen™564 in a thin layer at 40% humidity and 23°C.

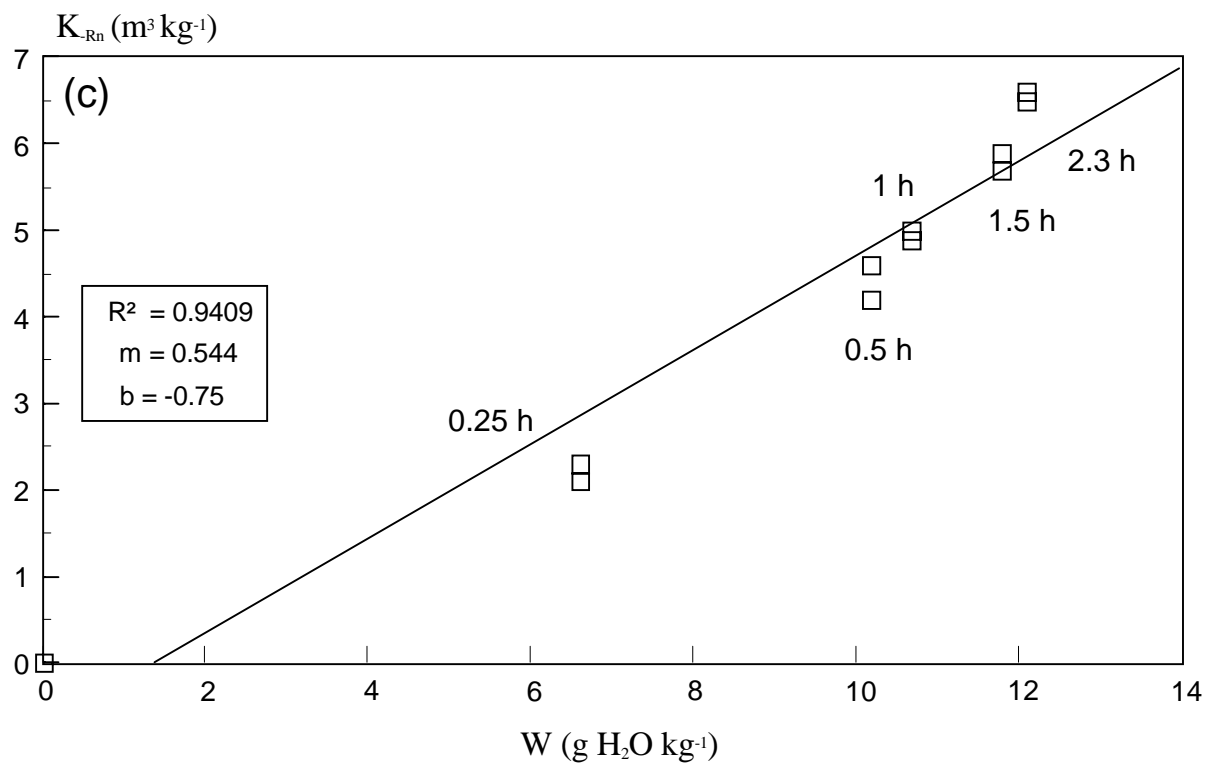
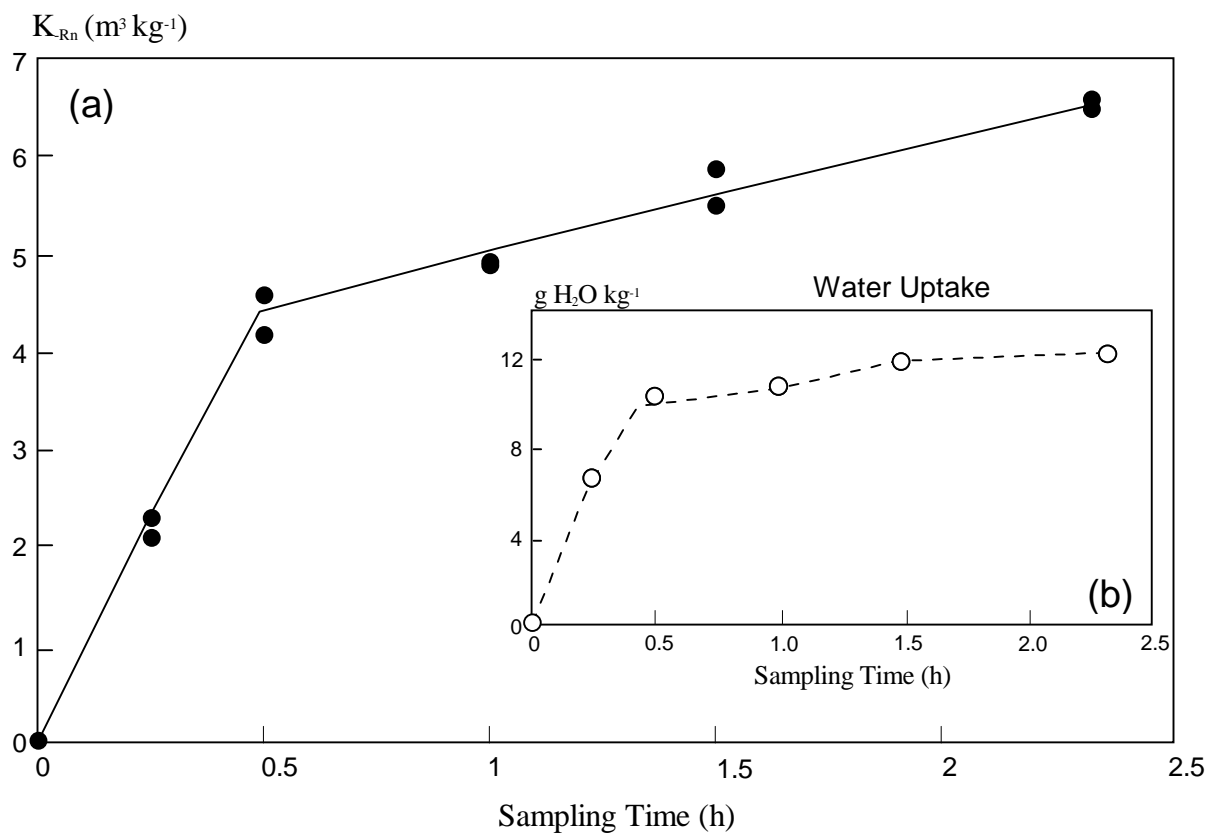


Figure 10a, b, c. ^{222}Rn and H_2O adsorption: 6 cm tubes of Carboxen™-564: sampling flow rate = $4.2 \times 10^{-3} \text{ m}^3 \text{ h}^{-1}$; $C_0 = 1260 \text{ Bq m}^{-3}$; $T = 25^\circ\text{C}$ and $\text{RH} = 29 \pm 5\%$.

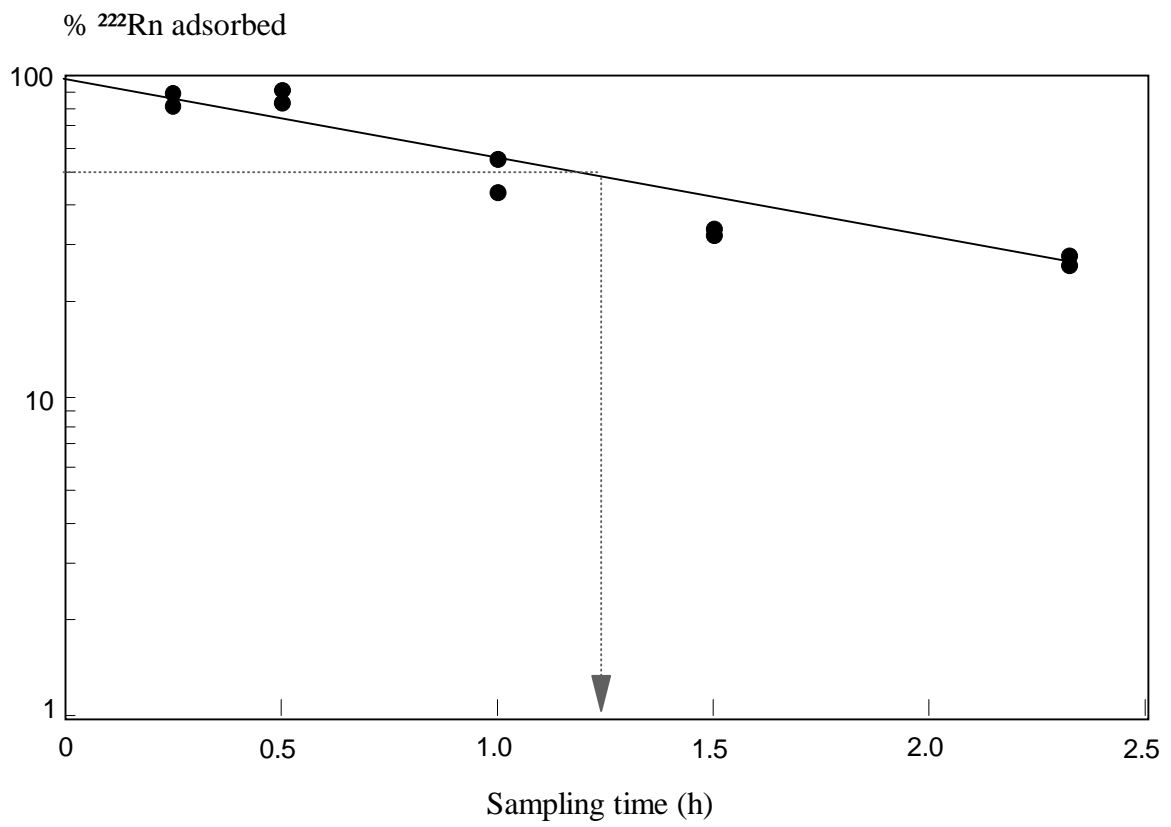


Figure 11. Collection efficiency of 0.4 g Carboxen™-564: sampling flow rate = $4.2 \times 10^{-3} \text{ m}^3 \text{ h}^{-1}$; $T = 25^\circ\text{C}$; $\text{RH} = 29\%$.

A PPENDIX

SORBENT BEHAVIOR IN A THIN LAYER AND A PACKED BED BCMS

Absolute humidity and air water vapor content

It was shown in Fig. 6a that adsorbed water vapor was the major interferant in the ^{222}Rn collection efficiency of the sorbent. Eqn 5a quantified the reduction in the % collection efficiency due to coadsorbed water vapor in the Polyport system. The amount of water vapor adsorbed by the sorbent will depend on the air water vapor content.

Fig. 7 shows the absolute humidity vs. temperature chart used to determine the air water vapor content, C_w (g $\text{H}_2\text{O m}^{-3}\text{-air}$), in Fig. 6b The linear log-log relationships in Fig. 7 were determined from data obtained in the Steam Tables, Handbook of Physics and Chemistry, and can be approximated by the following relationship for temperatures ranging from 5°C - 40°C :

$$C_w \text{ (g H}_2\text{O m}^{-3} \text{ air)} = 0.0615 \exp [0.0533 \times T \text{ (}^\circ\text{C)}] \times \text{RH (\%)} \quad (\text{A1})$$

Passive sampling of ^{222}Rn in a thin layer

Fig. 8 shows a log-log plot of water uptake vs. time for CarboxenTM-564, exposed as a thin layer to 40% and 67% RH at room temperature. The data show a nonlinear relationship between water uptake and time, and a factor of two reduction in the radon adsorption coefficient, K_{Rn} , as sorbent water uptake increases by a factor of 2, due to increased humidity.

Seven samples, containing gram amounts of oven-dried CarboxenTM-564, were aliquated into a 3.5-cm diameter by 1-cm high plastic containers. The sorbents were exposed in a thin layer to ^{222}Rn at 23°C and 40% RH and removed at fixed intervals over a 24-h period. $K_{\text{Rn}}(t)$ was then determined using eqn A2 (below) and is plotted in Fig. 9a.

$$K_{\text{Rn}}(t) \text{ (m}^{-3} \text{ kg}^{-1}\text{)} = Q_t C_o^{-1} M^{-1} \quad (\text{A2})$$

where M is the mass of CarboxenTM-564, kg.

The data in Fig. 9a show that the ^{222}Rn adsorption coefficient for CarboxenTM-564 in a thin layer achieved a value of $6.5 \text{ m}^3 \text{ kg}^{-1}$ in 24-h that is much higher than most activated charcoals. This is due primarily to a marked reduction in water vapor uptake (0.020 kg kg^{-1}) as compared with other activated charcoals (Scarpitta 1992). Fig. 9a also shows the ^{222}Rn removal from

CarboxenTM-564 following a 24 h exposure. Multiple gamma counts made using open containers of exposed sorbent over a 5-h period show that the ²²²Rn desorption time constant corresponds to 2.0 h.

The data in Fig. 9b represent the corresponding amounts of water vapor adsorbed during the exposure interval. The relationship between the amount of adsorbed water vapor, w , and K_{Rn} is linear over the 24-h exposure period (see Fig. 9c).

Effect of temperature on K_{Rn}

$K_{Rn}(t)$, determined experimentally from the amount of adsorbed ²²²Rn, is time, temperature and humidity dependent, and characterizes a sorbent's ability to adsorb ²²²Rn gas. The magnitude of K_{Rn} ($m^3 kg^{-1}$) represents the volume of the air sampled per unit weight of sorbent. Typically, its magnitude exceeds the limiting pore volume space of a sorbent, V_o ($m^3 kg^{-1}$). The maximum amount of ²²²Rn adsorbed by a dry sorbent, expressed in terms of the ²²²Rn adsorption coefficient, temperature and the available pore volume space has been derived from the Polanyi based Dubinin-Radushkevich equation (Scarpitta 1995)

$$\text{Ln } K_{Rn,T}^o - \text{Ln } V_o = V_r E_{Rn} [RT]^{-1} \quad (\text{A3})$$

where V_o = the apparent limiting pore volume of sorbent, $m^3 kg^{-1}$; E_{Rn} = the potential energy for ²²²Rn adsorption in the absence of water vapor, $J m^{-3}$; R = the ideal gas constant, $8.314 J mol^{-1} K^{-1}$; T = absolute temperature, $^{\circ}K$; V_r = molar volume of ²²²Rn ($50.45 \times 10^{-6} m^3 mole^{-1}$). The molar volume is the ratio of molecular weight to density at the boiling point. Assuming that the equilibrium $K_{Rn,T}^o$ value for CarboxenTM-564 at $25^{\circ}C$ ($298^{\circ}K$; dry air) is $7.0 m^3 kg^{-1}$, and $V_o = 1 \times 10^{-3} m^3 kg^{-1}$, then eqn A3 reduces to

$$\text{Ln } K_{Rn,T}^o + 6.9 = V_r E_{Rn} [RT]^{-1} \quad (\text{A4})$$

from which E_{Rn} is $434.64 \times 10^6 J m^{-3}$.

Equation A4 can be resolved for $K_{Rn,T}^o$ at different values of T ($^{\circ}K$) with E_{Rn} , V_r and R as constants. The values for $K_{Rn,T}^o$ are 9.5 and $5.3 m^3 kg^{-1}$ at $15^{\circ}C$ ($288.15^{\circ}K$) and $35^{\circ}C$ ($308.15^{\circ}K$), respectively. This shows that the magnitude of $K_{Rn,T}^o$ decreases with increasing temperature, and there should be at least a 20% difference in the magnitude of $K_{Rn,T}^o$ (and so, the % collection efficiency) over a $10^{\circ}C$ range in the absence of water vapor.

Dynamic sampling : packed bed configuration

Fig. 10a shows the results obtained when ^{222}Rn laden air was passed through five pairs of tubes containing 0.4 g of the BCMS material at a sampling flow rate of 4.2 L h^{-1} . The amount of ^{222}Rn , Q_t (Bq), adsorbed by the BCMS material was determined by LS counting, and was used in eqn 7 to determine the dynamic ^{222}Rn adsorption coefficients, K'_{Rn} .

The data show that ^{222}Rn build up in a 6-cm tube is similar in profile to that of the same sorbent maintained as a thin layer for 24 h (see Fig. 9a). K'_{Rn} corresponds to $6.5 \text{ m}^3 \text{ kg}^{-1}$ after 2.3 h of sampling. Water vapor adsorption also exhibited a profile similar to that of a thin layer, achieving a maximum value of 12.2 g kg^{-1} at 29% RH (see Fig. 10b).

The same data are presented in Fig. 11 as a semi-log plot of the ^{222}Rn collection efficiency vs. sampling time. The ^{222}Rn collection efficiencies were determined from Q_t (Bq), using eqns 1 - 3. The data show a marked decrease in the ^{222}Rn collection efficiency with increasing sampling time when the sampling flow rate was maintained at 4.2 L h^{-1} . The half-time ^{222}Rn retention for the sorbent tube, which corresponds to a 50% breakthrough, was 1.2 h. This value, multiplied by 1.44, agrees with the measured desorption time-constant value of 2 h from a thin layer of material. At 50% retention, K_{Rn} was estimated to be $5.0 \text{ m}^3 \text{ kg}^{-1}$. From the data in Fig. 10a and eqn 7, the amount of ^{222}Rn adsorbed by the sorbent at the 50 percentile is estimated to be about 3 Bq when the ambient ^{222}Rn concentration was 1260 Bq m^{-3} . A ^{222}Rn activity of 3 Bq (180 dpm) can be measured by either gamma or LS counting. A two-order of magnitude reduction in the ambient ^{222}Rn concentration to 10 Bq m^{-3} suggests that the total amount of ^{222}Rn adsorbed by the sorbent at a sampling flow rate of 5 L h^{-1} will be about 30 mBq which is below the LLD of the gamma detecting instrument, but above that of the LS counting instrument.



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